ULTRASONIC MEASUREMENTS OF ELASTIC CONSTANTS IN POLYMERIC MATRIX/GLASS FIBERS COMPOSITE MATERIALS VERSUS TEMPERATURE

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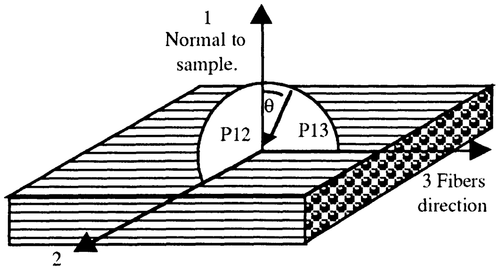
INTRODUCTION

There are many methods to measure the properties of composite materials. For instance, the elastic constants of orthotropic composite materials can be identified from the measurement of waves velocities by cutting the samples in various directions to access to all constants [1] . These destructive measurements are not available when the purpose is to follow the evolution of materials after or during the exposure of a damage (stress, corrosion, temperature ...). Composite materials are often made of the plies superposition in the shape of thin plates [2]. So, the well-known immersion method [3,4,5 , 6 ...] is very well adapted to the non-destructive evaluation of plate shaped, orthotropic materials.
In this paper, we present an extension of this method to follow the stiffness tensor evolutions when the temperature approaches the glass transition temperature.
First, it is interesting, from an engineering point of view, to know the elastic behavior of composite materials versus temperature. The second aim of this method, is to let us easily change the properties of one of the material constituents. Then micromechanical models to predict elastic moduli can be compared to the measurement, and validated or refuted.
Results are given for neat epoxy matrix and unidirectional glass fibers composite. The quality of measurements is tested by comparison with classical prediction model. It is verified that not only the initial properties are anisotropic, but also their variations.



$$C_{ij} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ & C_{22} & C_{23} & 0 & 0 & 0 \\ & & C_{33} & 0 & 0 & 0 \\ & & & C_{44} & 0 & 0 \\ & \text{Sym} & & & C_{55} & 0 \\ & & & & & C_{66} \end{bmatrix}$$

Figure 1. Definitions of coordinates system and tensor of elasticity.

IMMERSION METHOD TO MEASURE ELASTIC CONSTANT VERSUS TEMPERATURE

The existence of three mutually orthogonal planes of symmetry imposes the orthotropic class of symmetry [7], and the corresponding elasticity tensor $\left[C_{ij}\right]$ with nine independent constants is given at figure 1. This preliminary study is limited to unidirectional fibers composites. So, there are only five independent constants [7] C_{11}, C_{66}, C_{33}, C_{55}, and C_{13} to describe the composite elasticity. The small plate-shaped sample is immersed between immersion broadband transducers and sustained with a goniometer to impose the angle of incidence θ. The measurements of velocities versus θ in the planes of symmetry P_{12} and P_{13} (Fig.1), lead to the identification of C_{ij} in the planes of incidence [4..6]. To reach the temperatures for which the constants are changing (≈ 100°C to 150°C), we used broadband (≈ 1 to 4 MHz), commercial transducers sustaining temperatures up to 250°C. A low viscosity silicone oil is the coupling medium for temperatures higher than 70°C.
The wave velocity in water versus temperature is well known, but the wave velocity in oil needs to be measured.

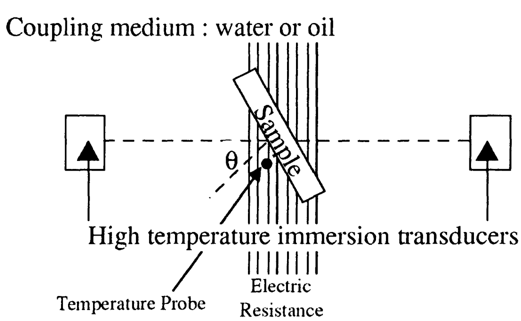


Figure 2. High temperature, immersion ultrasonic system.

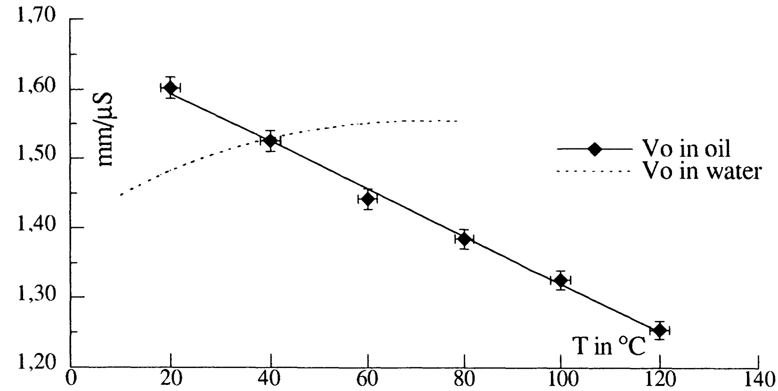


Figure 3. Evolution of wave velocities in water and oil versus temperature.

The velocity V in the sample, for a given angle θ, is obtained from the difference in times of flight Δt between the waveform transmitted without sample (reference) and with sample, by the equation :

$$V = V_0 / \sqrt{1 + a(a - 2\cos(\theta)} \ , \tag{1}$$

where $a = V_0 \Delta t / e$, e is the sample thickness and V_0 the coupling medium velocity. When measuring V, V_0 is a known quantity. The equation (1) can also be used to measure V_0, if V is known. First we measured the velocity of a glass sample immersed in water as coupling medium and verified that the properties of glass are constant in the range of temperatures 10..70°C. Then the glass sample was immersed in oil and V_0 was measured versus temperature using equation (1). The best fitting of the experimental values of velocities in oil (Fig.3) gives the expression :

$$V_0 = 1.662 - 0.00342 \ T \ \text{mm}/\mu\text{S} \ . \tag{2}$$

This linear evolution is used with equation (1) to measure the samples velocities versus the temperature, when they are immersed in oil. The system was tested by measuring the glass velocities in the range of temperatures 20..120°C. Figure 4 gives the actual errors. The mean values of the longitudinal and shear velocities (Cl = 5,86 ± 0,02 mm/µS and Ct = 3,28 ± 0,05 mm/µS) were found stable enough to start using the system. The sample and coupling medium velocities do not rapidly change with temperature, but there is a real difficulty to measure precisely the velocity with this substitution method. The time of flight (TOF) between transducers is around 100 µS. The perturbation introduced by the sample is about 2 µs. For an acceptable measurement, the relative TOF stability needs to be about 10^{-4} which corresponds to a temperature stability of 0.1 °C along the waves path. This is not currently achieved and is the main source of errors.

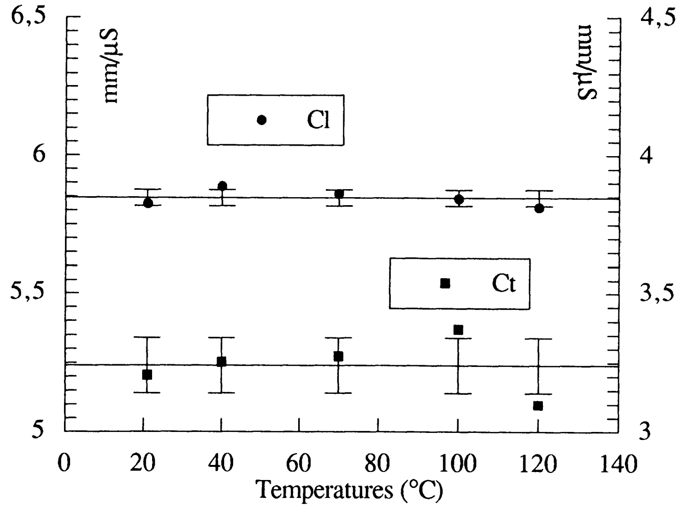


Figure 4. Velocities in glass versus temperature.

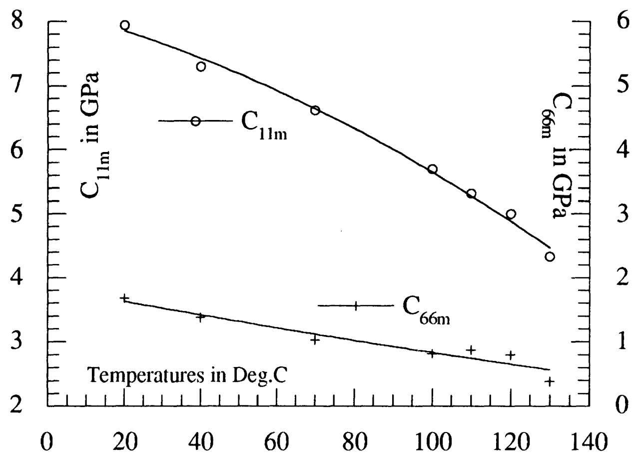


Figure 5. Elastic properties of neat matrix versus temperature.

RESULTS

The method is tested on a material made of epoxy matrix reinforced with long unidirectional E-glass fibers. The composite sample has 32 plies. The mass density is 1.79 gr/cm^3, the thickness is 5.72 mm and the volume fraction of fibers is $V_f = 0.448$.

To predict the elastic constants of the composite, a sample made of neat matrix was cured in the same conditions and it is supposed to have the same properties as the composite matrix.

The best and simplest models for the variations of the matrix elastic properties with temperature are optimized from the measurements (Fig.5) and give two equations that are used to predict the elastic properties of the composite :

$$C_{11m} = 8.19 - 0.015\,T - 0.00011\,T^2 \text{ GPa and } C_{66m} = 1.81 - 0.0096\,T \text{ GPa}. \quad (3)$$

Figure 6 shows the velocities in the plane P_{13} for the two extreme temperatures 20°C and 130°C. Clearly, the relative changes of the longitudinal properties are greater in the direction 1 (perpendicular to the fibers)than in the direction 3 (along the fibers). This is easily explained by the fact that the longitudinal properties of the material in direction 1 are essentially controlled by the matrix, and by the fibers in direction 3.

COMPARISON WITH MICROMECHANICS MODELS

The equations (3) giving the matrix properties, are included in a micromechanic model to predict the properties of the composite materials versus the temperature and to compare them with the experimental data. The E glass fibers characteristics can be found in the literature $C_{11f} = 90$ GPa and $C_{66f} = 30$ GPa. The fibers volume fraction is $V_f = 0.448$ and $V_f = 1 - V_m$.

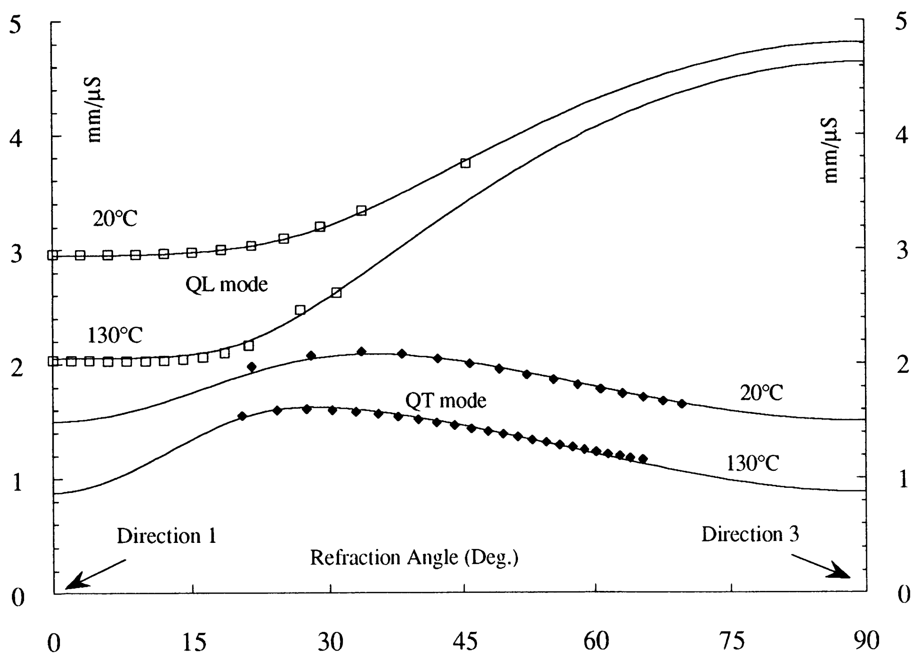


Figure 6. Velocities in the plane P_{13} for two temperatures 20°C and 130°C.
scattered points : measured velocities
solid lines : computed velocities from C_{ij}

To adapt the Hashin's [8,9] model, based on engineering constants, to the elastic constants, the Coulomb's moduli G_f, G_m, bulk moduli k_f, k_m, Young's moduli E_f, E_m and the Poisson's ratios ν_f, ν_m of fibers and matrix are easily obtained from the equations :

$$G = C_{66}, \qquad k = C_{11} - C_{66}, \qquad E = \frac{C_{66}(3C_{11} - 4C_{66})}{(C_{11} - C_{66})}, \qquad \nu = \frac{(C_{11} - 2C_{66})}{2(C_{11} - C_{66})} \tag{4}$$

For unidirectional composites, the plane strain bulk modulus k, the Young's modulus E_l in fibers direction, the axial shear modulus G_l and the axial Poisson's ratio ν_l are given [8] by :

$$k = k_m(k_f + G_m)V_m + k_f(k_m + G_m)V_f / ((k_f + G_m)V_m + k_f(k_m + G_m)V_f) \tag{5}$$

$$G_l = G_m(G_m V_m + G_f(1 + V_f)) / (G_m(1 + V_f) + G_f V_m) \tag{6}$$

$$E_l = E_m V_m + E_f V_f + 4V_m V_f(\nu_f - \nu_m)^2 / \left({}^{V_m}\!/_{k_f} + {}^{V_f}\!/_{k_m} + {}^{1}\!/_{G_m} \right) \tag{7}$$

$$\nu_l = \nu_m V_m + \nu_f V_f + V_m V_f(\nu_f - \nu_m)\left({}^{1}\!/_{k_m} - {}^{1}\!/_{k_f} \right) / \left({}^{V_m}\!/_{k_f} + {}^{V_f}\!/_{k_m} + {}^{1}\!/_{G_m} \right) \tag{8}$$

The transverse shear modulus G_t is equalled to its lower bounds [9] by :

$$G_t = G_m + \frac{V_f}{1/(G_f - G_m) + (k_m + 2G_m)V_m / 2(k_m + G_m)G_m} \tag{9}$$

From the preceding equations linking matrix and fibers properties to the composite properties, it is easy to predict the five independent composite elastic constants from the matrix and fibers properties :

$$C_{11} = k + G_t \quad C_{66} = G_t \qquad C_{13} = 2\nu_l k \qquad C_{33} = 4\nu_l^2 k + E_l \qquad C_{55} = G_l \tag{10}$$

The other constants of a transverse isotropic material are given by :

$$C_{22} = C_{11} \qquad C_{12} = C_{11} - 2C_{66} \qquad C_{44} = C_{55} \qquad C_{23} = C_{13} \tag{11}$$

Figures 7 and 8 show the comparisons between the measurements of C_{ij} and their predictions. Formula (9) associated with the lower bound of C_{66}, seems well correlated with measurements. The largest discrepancy is related to the constant C_{13}, for which we do not have any explanation now.

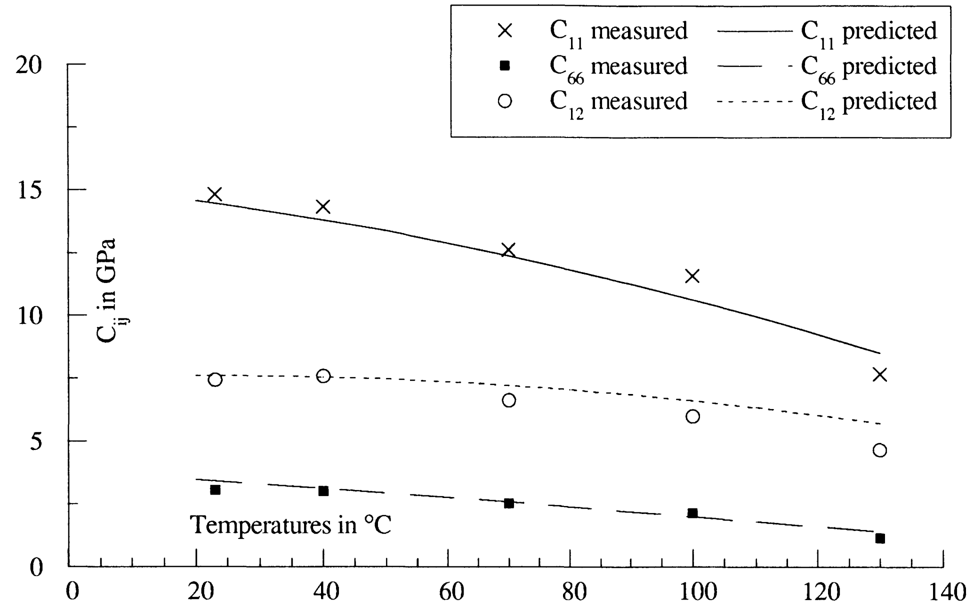


Figure 7. Comparison between measured and predicted C_{ij} in the isotropic plane P_{12}.

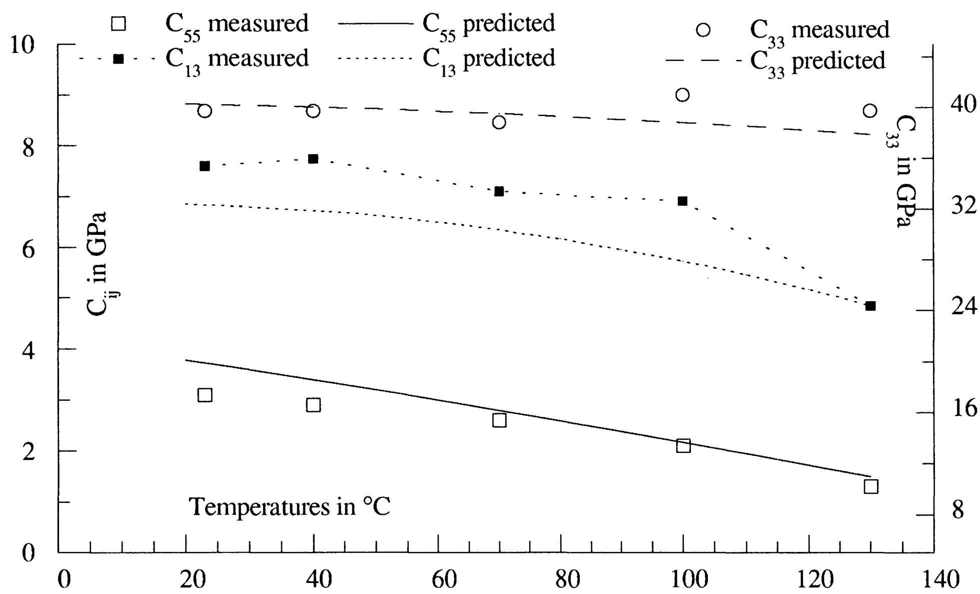


Figure 8. Comparison between measured and predicted C_{ij} in the anisotropic plane P_{13}.

CONCLUSIONS

This paper reports on preliminary measurements of elastic constant evolution of epoxy matrix/ glass fibers composite materials versus temperature. The main cause of measurement errors is identified and the temperature stability of the system is under way.

However, the elastic constant modifications are well correlated with the predicted ones by micromechanic models.

Some other causes of errors are still difficult to detect. We need to find a way to be sure that the neat matrix properties are the same than the composite matrix. The fibers properties can be slightly different than those found in the literature. In the next steps of that work, we will try to elucidate those questions, before studying the complex stiffness [10] tensor versus temperature.

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REFERENCES

1. R.D.Kritz and W.W.Stinchcomb, "Elastic Moduli of Transversaly Isotropic Graphite Fibers and Their Composites" Exp.Mech. 19, 41-49 (1979).
2. S.W.Tsai- "Composites Design." Think Composites, Dayton, Paris, Tokyo (1987).
3. R.Kono "The dynamic bulk viscosity of polystyrene and polymethyl methacrylate" J.Phys.Soc.Japan 15 (4) 718-725 (1960)
4. B.Hosten, A.Barrot, J.Roux- "Interférométrie Numérique Ultrasonore pour la détermination de la Matrice de Raideur des Matériaux Composites." Acustica. 53 212-217 (1983).
5. S.I.Rokhlin and W.Wang, "Critical Angle Measurement of Elastic Constants in Composite Material",J. Acous. Soc. Am., 86, (5) , 1876-1882 (1989).
6. B.Hosten "Elastic Characterization of Orthotropic Composite Materials from Ultrasonic Inspection through non Principal Planes." Review of progress in QNDE, Vol. 10b 1437-1444 (1991).
7. B.A.Auld "Acoustic fields and waves in solids." Robert E.Krieger Publishing Company, Malabar,Florida (1990).
8. Z. Hashin. "Complex Moduli of Viscoelastic Composites-II.Fiber Reinforced Materials" Int.J.Solids Structures, 6 pp 797-807 (1970).
9. Z. Hashin. "Analysis of Composite Materials. A Survey." Journal of Applied Mechanics 50 481-504 (1983).
10. B. Hosten. "Reflection and Transmission of Acoustic Plane Waves on an Immersed Orthotropic and Viscoelastic Solid Layer." J. Acous. Soc. Am. 89 2745-2752 (1991).